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Silicone elastomers with aromatic voltage stabilizers

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Abstract

A method to synthesise a polydimethylsiloxane-polyphenylmethylsiloxane (PDMS-PPMS) copolymer with electron-trapping effects has been developed utilising a quick hydrosilylation reaction at ambient temperature and crosslinking via hydrosilylation by a vinyl-functional crosslinker. The mechanism of electron-trapping by aromatic compounds grafted to silicone backbones in a crosslinked PDMS is illustrated in Fig. 1. The electrical breakdown strength, the storage modulus and the loss modulus of the elastomer were investigated, as well as the excitation energy from the collision between electron carriers and benzene rings in PDMS-PPMS copolymer was measured by UV-vis spectroscopy. The developed elastomers were inherently soft with enhanced electrical breakdown strength due to delocalized pi-electrons of aromatic rings attached to the silicone backbone. The dielectric relative permittivity of PDMS-PPMS copolymers remained between 2 to 3 with low conductivity and low dielectric loss as well as high storage moduli with low viscous loss, thereby maintaining the electro-mechanical integrity of the elastomer.

Motivation: Increasing the electrical breakdown strength of DEs allows for larger actuation due to the possibility of utilizing larger electrical fields.^{1,2} Many studies on electrical breakdown of pre-strained DEs have been conducted, however less efforts have been focused on improving the electrical breakdown strength such as by blending in additives or by polymer structure modifications. In order to increase the electrical breakdown strength of polymers for e.g. the cable industry, additives like aromatic voltage stabilizers are used. Earlier works on using voltage stabilizers in polymers have mainly focused on polyethylene with the purpose of reducing power loss for high voltage insulation cables.³⁻⁵ As an alternative to utilise additives as voltage stabilizers, grafting aromatic compounds to silicone backbones may overcome the common problem of insolubility of the aromatic voltage stabilizer in the silicone elastomers due to phase separation. Preventing phase separation during preparation and during operation is a key requirement for long life-times of DEs.⁵

Introduction

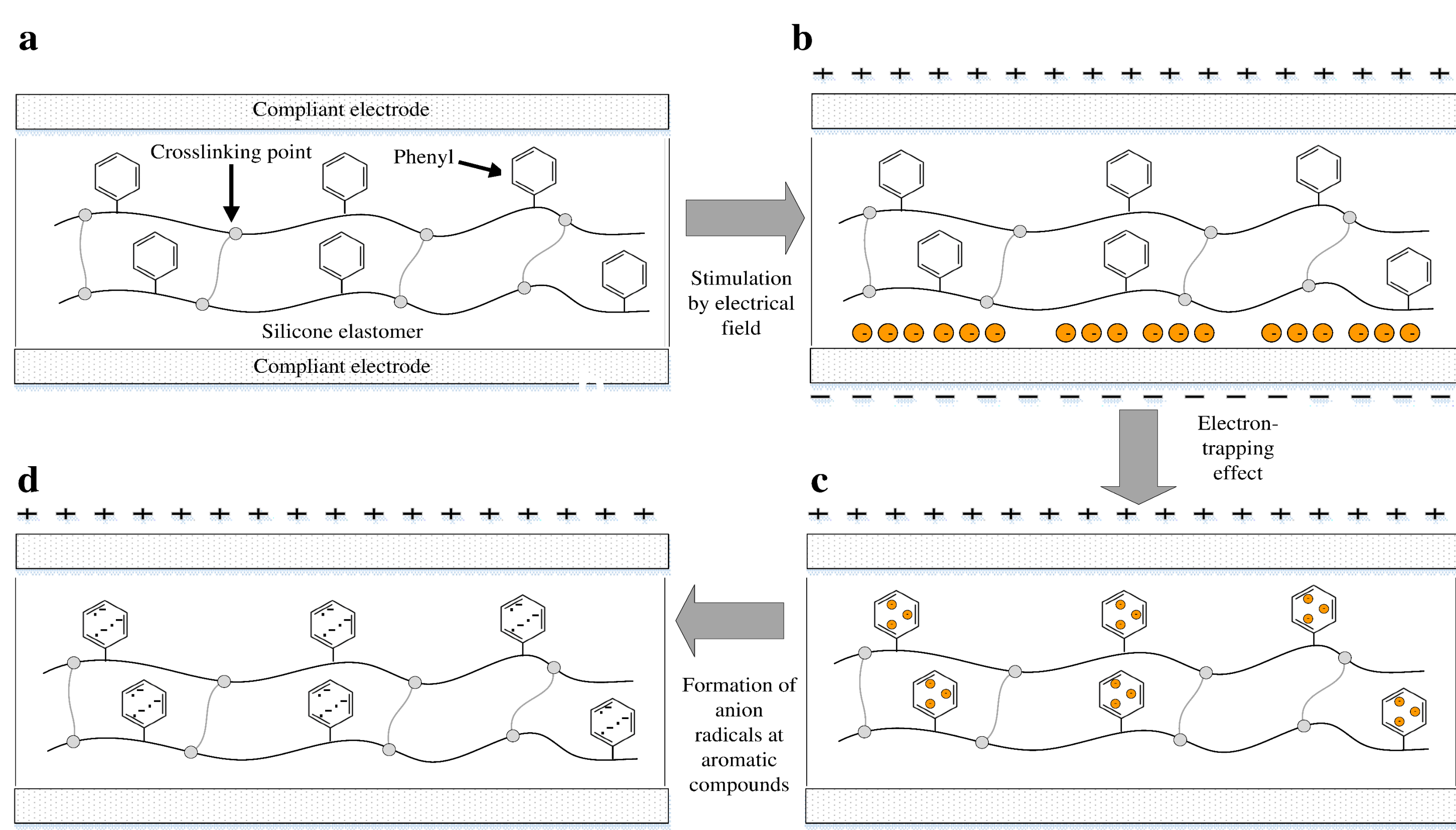
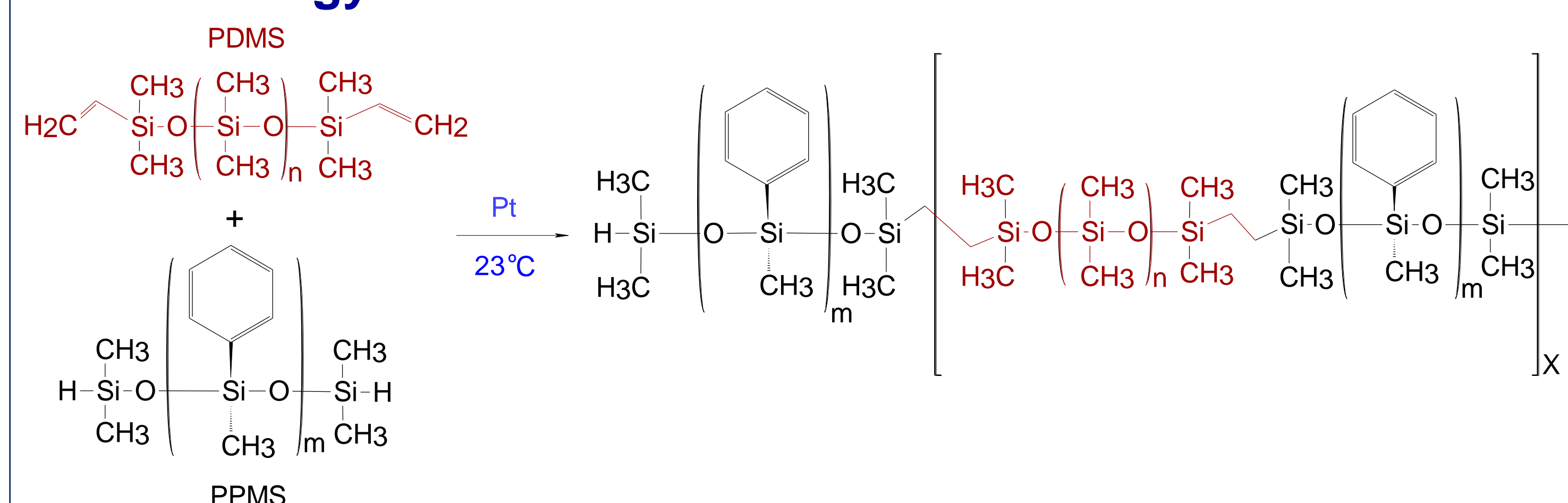


Fig. 1 A mechanism of electron-trapping: **a)** A silicone elastomer with aromatic compounds attached to silicone backbone and sandwiched between two compliant electrodes. **b)** The existence of electrons at the interfaces between the elastomer and the compliant electrode in a presence of electrical field. **c)** The electron-trapping effect as a consequence of collision between electrons and aromatic rings. **d)** The formation of anion radicals resulted from disturbance of the cloud of π -electrons.

Methodology



Scheme 1 The hydrosilylation reaction utilized when preparing PDMS-PPMS block copolymer with stoichiometric ratio of $r = (X+1) / X$, where $m = 2, 6$ are numbers of repeating phenylmethylsiloxane units in PPMS, and $n = 377, 231, 126, 80$ are numbers of repeating dimethylsiloxane units in PDMS.

Results

a) Electrical breakdown strength

Table 1 Dielectric breakdown strength at 23 °C, Weibull parameters α and β , and R^2 of linear fit for the reference elastomer (DMS-H31) and crosslinked PDMS-PPMS copolymers with different concentration of aromatic compounds.

Crosslinked PDMS-PPMS copolymer	Thickness (μm)	Electrical breakdown strength ($\text{V}/\mu\text{m}$)	Weibull β -parameter	Weibull η -parameter	R^2	Normalised electrical breakdown ($\text{V}/\mu\text{m}$)
DMS-H31 (ref.)	105	53 \pm 4	17	55	0.85	52.9
377DMS_2PMS	81	53 \pm 4	17	55	0.85	53.7
231DMS_2PMS	91	60 \pm 4	20	61	0.91	60.1
126DMS_2PMS	80	65 \pm 2	32	66	0.94	65.5
80DMS_2PMS	114	54 \pm 7	20	53	0.90	53.6
377DMS_6PMS	81	64 \pm 2	47	65	0.89	64.1
231DMS_6PMS	95	54 \pm 1	60	54	0.94	53.6
126DMS_6PMS	95	54 \pm 2	39	55	0.88	54.0
80DMS_6PMS	95	56 \pm 2	28	57	0.94	56.1

b) Weibull analysis

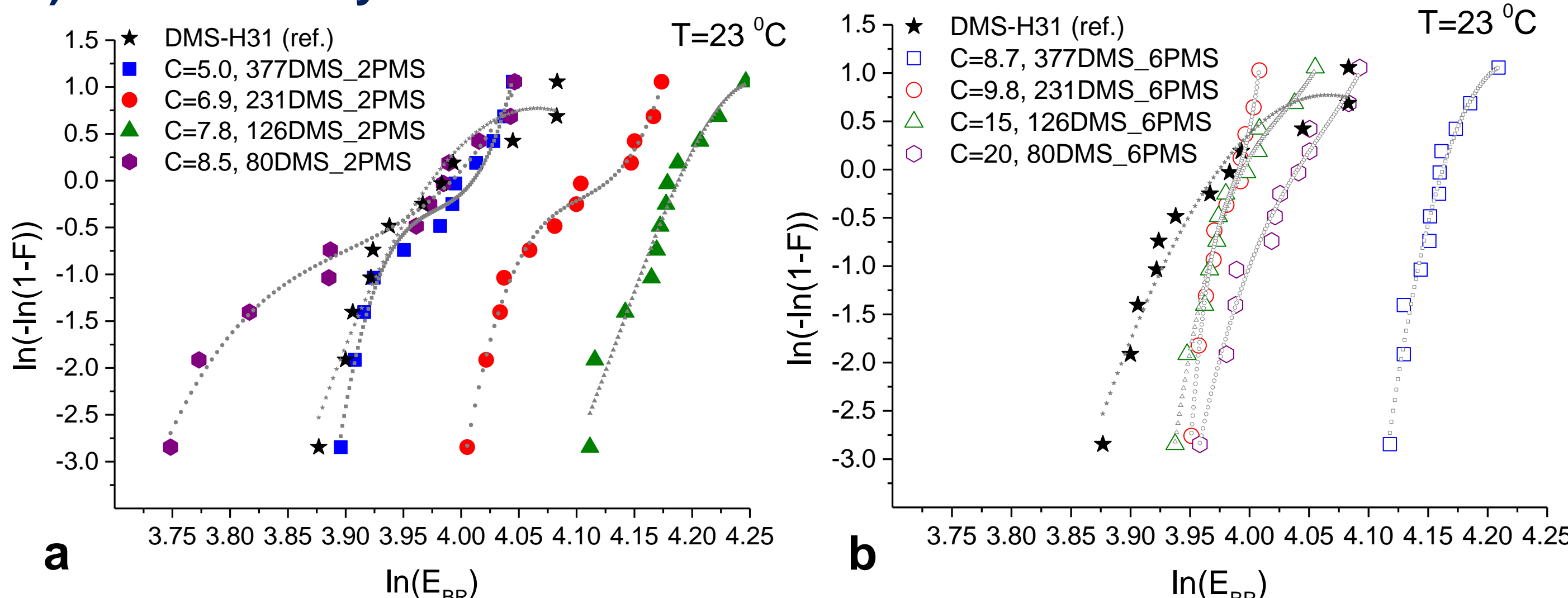


Fig. 2 Cumulative probability of failure of PDMS elastomer and PDMS-PPMS copolymers with different concentration of aromatic compounds: **a)** The copolymers with short-chain PPMS, **b)** The copolymers with long-chain PPMS. The dashed lines are the guidelines for eyes to observe and differentiate slopes of the data; C is in $10^{-4} \text{ mol g}^{-1}$.

c) Conductivity

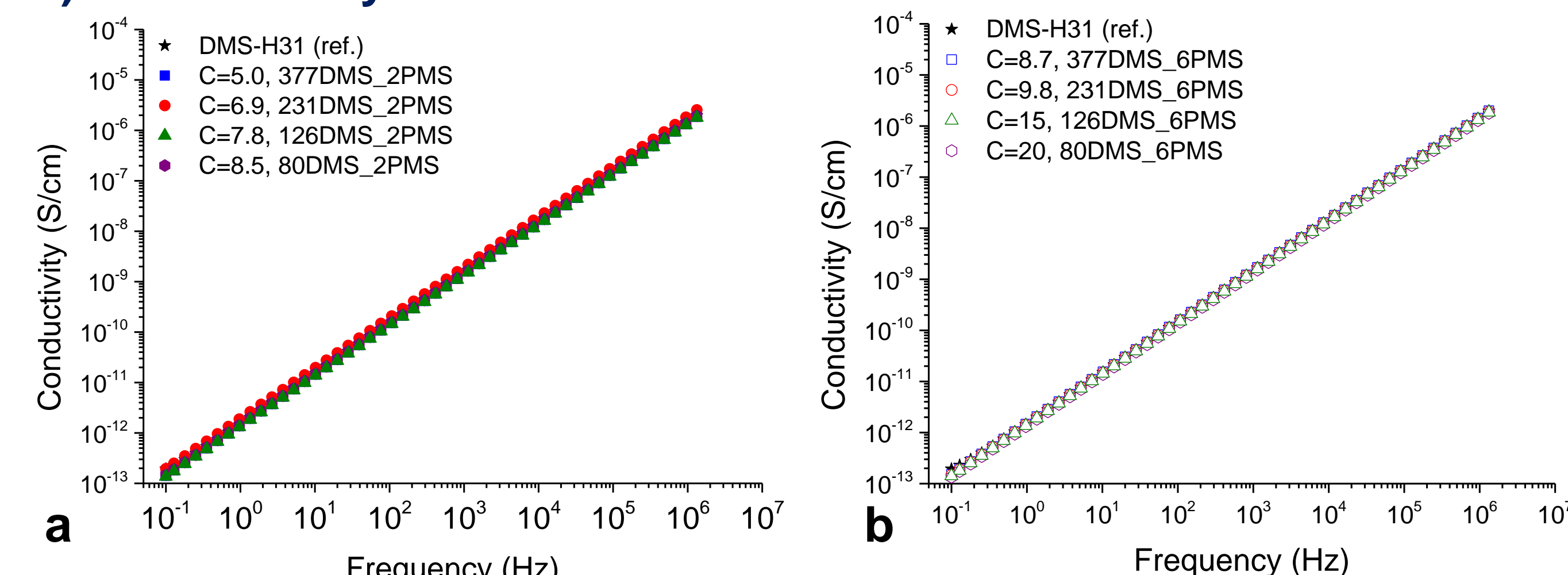


Fig. 3 The conductivity of PDMS-PPMS elastomers with different concentrations of aromatic compounds at 23 °C: **a)** short-chain PPMS, **b)** long-chain PPMS block; C is in $10^{-4} \text{ mol g}^{-1}$.

d) Mechanical properties and stress-strain behaviour

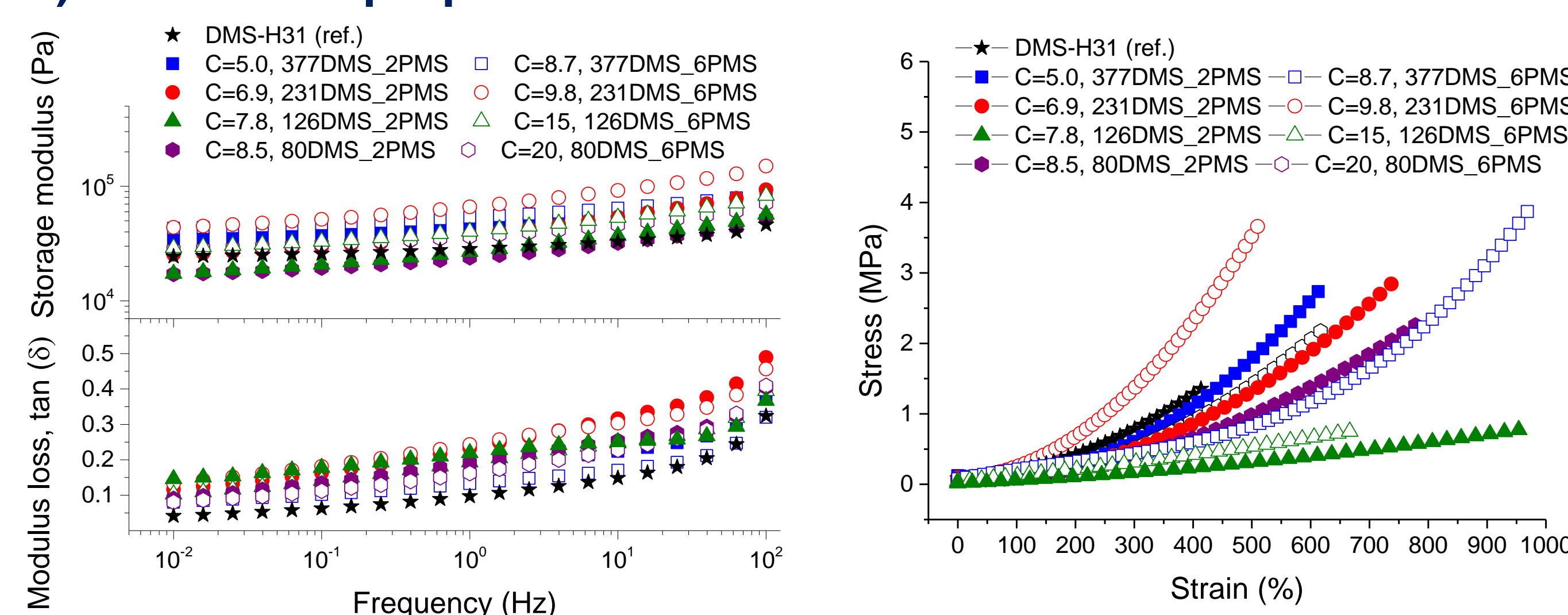


Fig. 4 The storage and loss moduli of PDMS-PPMS elastomers with different concentrations of aromatic compounds at 23 °C; C is in $10^{-4} \text{ mol g}^{-1}$.

Conclusion

- Aside of having high electrical breakdown strength, the prepared PDMS-PPMS elastomer with different concentration of aromatic compounds showed increased storage modulus with low viscous loss as well as increased strain at break, hence maintaining electro-mechanical integrity of the elastomer.
- The increased Young's moduli increases mechanical stability with strain-hardening behaviour, which is a desirable property for dielectric elastomers due to the suppression of electromechanical instability for long life-times.

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